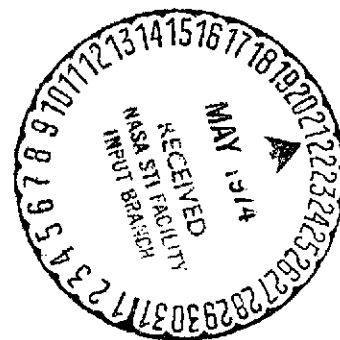


MAGNETO-OPTICS.
OPTICAL DETECTION OF PARAMAGNETIC ELECTRONIC RESONANCE
BY MEASURING THE PARAMAGNETIC ROTARY POLARIZATION OF A VISIBLE RADIATION

Alfred Kastler

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It is demonstrated that the electronic magnetic resonance of paramagnetic ions produced by a high-frequency magnetic field in the presence of a constant field must influence the magnetic rotary polarization of these ions observed with a visible radiation.

Paramagnetic electronic resonance in the radio frequency region was predicted by Dorfmann in 1923 [1] and discovered by Zavoisky in 1945 [2] following the work of Gorter et al. [3] on paramagnetic relaxation. Since 1946, it has been the object of numerous investigations [4].

In the majority of the investigations on this phenomenon, the resonance is produced and detected by radioelectric methods. However, it might be interesting to use other effects for detection: magnetomechanical, calorific, or optical. The resonance transitions produce a modification in the populations of the magnetic sublevels m of the fundamental state of the ions, populations which are governed by Boltzmann's law at thermal equilibrium. Every phenomenon which depends on the distribution of elementary magnets between the different m levels could thus serve to detect the transitions which change the respective populations of these levels.

Bitter has drawn attention to the advantage of optical detection methods, and, especially, measurement of the polarization of optical resonance radiations has been proposed and applied to the study of the resonance of excited atomic levels [5].

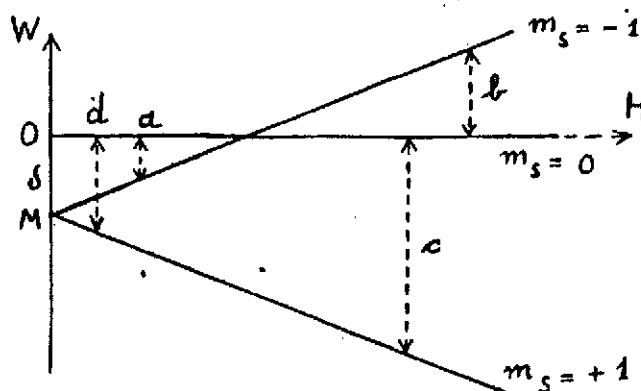
Another optical effect which depends directly on the distribution of ions among the m levels of the fundamental state is paramagnetic rotary polarization. In crystals or in paramagnetic solutions, the Faraday effect has a double origin: 1) the diamagnetic effect, common to all bodies, a consequence of Larmor precession; 2) the paramagnetic effect, discovered by Jean Becquerel, which is due to the orientation of elementary magnets by the field which induces an asymmetry of the intensity of the Zeeman absorption components. This second effect follows a Curie law at $1/T$ and shows a saturation at low temperature. The rotary polarization of paramagnetic ions and its variation as a function of temperature have been the object of systematic investigations by Jean Becquerel et al. [6]. The theory of the phenomenon has been elaborated by many authors [7].

One can predict the conditions for which the effect sought is likely to be maximum. The use of a low temperature is favorable for two reasons: the asymmetry of orientation and the value of the rotary polarization are large, and the relaxation times of the "magnetic-network levels" are long. A long relaxation time facilitates the saturation obtained with a given radioelectric power. Complications introduced by the crystalline field must also be taken into account. For ions of rare earths, the crystalline field produces separations on the order of 20 to 200 cm^{-1} . The quantum number j preserves its signification, and the levels separated by the field are the levels $|m_j|$. Degenerations which exist depend on the symmetry of the field, and if j is a half-integral, there is always Kramers degeneration. The levels $m_j = +1/2$ and $m_j = -1/2$ in particular are separated only by an exterior magnetic field. Thus, for ions where j is a half-integral, the resonance frequency which satisfies the selection rule $\Delta m_j = \pm 1$ is always that of the radioelectric region and must be reflected on the magnitude of the magnetic rotary polarization.

In the case of ions of the iron group, the crystalline field breaks the bond between the vectors \vec{L} and \vec{S} , the crystalline separations corresponding to variations in the infrared or visible region within the m_L levels, and to variations in the radioelectric region within the m_S levels. If one assumes that the rules of selection and polarization for optical transitions are the same as in the Paschen-Back effect ($\Delta m_L = \pm 1$ or $0, \Delta m_S = 0$), one is tempted to conclude that the radioelectric resonances $\Delta m_S = \pm 1$ must have no influence on the optical properties [8]. But studies on the magnetic rotary polarization of these ions show that the rotation is appreciable and that it depends essentially on the relative growth of the $\pm m_S$ levels.

A very interesting case is that of hexahydrated nickel fluosilicate whose magnetic rotary polarization [9] and magnetic resonance [10] have been studied. The figure on the following page shows the arrangement of the m_S levels of its fundamental state. From its law of variation as a function of temperature, magnetic rotary polarization is proportional to the difference in growth between the levels $m_S = -1$ and $m_S = +1$.

Examination of the figure shows that the resonance a should increase rotation, resonances b and c should decrease it, and, finally, resonance d



should reverse the sign of rotation if it approaches saturation. It would be interesting to submit these forecasts to the verification of experiment.

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